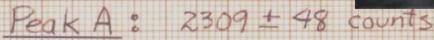
LOS ALAMOS NATIONAL LABORATORY

ACTINIDE RESEARCH **Q**UART

Minutes

AL GHIORSO RECALLS A MOMENT IN TIME



3.120 ± .065 c/m

Bkgnd= 0.129 ± .009

2991 ± .066 9m Net

Peak B:

161 ± 13 counts

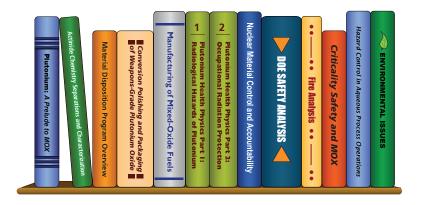


3.76 ± 0.64

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NRC OVERSEES LICENSING OF MOX FACILITIES

LOS ALAMOS DEVELOPS SEMINAR FOR NUCLEAR REGULATORY COMMISSION SAFETY REVIEWERS

A chance discussion has led to the development of a custom-designed seminar on mixed-oxide (MOX) fuels for members of the Nuclear Regulatory Commission (NRC). During a meeting several years ago between Vanice Perin of the NRC and Karl Staudhammer of what was then the Nuclear Materials Technology (NMT) Division at Los Alamos National Laboratory, the topic of training needs for the NRC came up. In particular, they discussed the need for NRC staff to acquire a broad yet strong knowledge of mixed oxides because of the resurgent interest by the United States in using excess plutonium from the nuclear stockpile for MOX fuel to generate electricity in commercial power plants.

The NRC, which was established after the Atomic Energy Agency was abolished by the Energy Reorganization Act of 1974, has regulatory responsibility for nuclear energy and nuclear safety in the U.S. Its regulatory

activities are focused on reactor-safety oversight and reactor licensing and renewal of existing plants, materials safety and licensing, and high- and low-level waste management. The NRC's Office of Nuclear Material Safety and Safeguards is responsible for licensing the MOX fuel fabrication facility, and the Office of Nuclear Reactor Regulation is responsible for licensing the use of the MOX fuel in commercial power plants.

Los Alamos was a natural fit to take the lead in the development of the seminar because of its 25-year history of working with oxide and mixed-oxide fuels, as well as its already established courses in actinide chemistry, handling of radiological materials, plutonium metallurgy, and criticality. Staudhammer and Randy Erickson, also of NMT Division, with input from the NRC, designed a seminar to meet the needs of safety reviewers to improve their understanding of production and operations relative to permitting issues for the approval of a MOX facility in the United States.

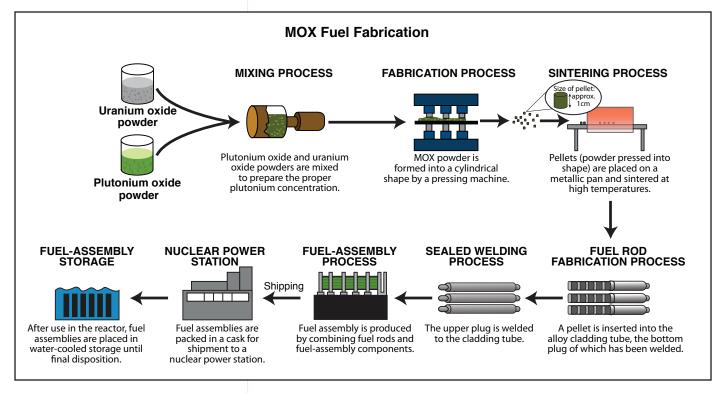
Background on MOX

The MOX program at Los Alamos is part of a much bigger picture in the U.S. effort to deal with 34 tons of surplus weapons-grade plutonium. The United States and Russia—under an arms-reduction treaty—are carefully planning to use the surplus plutonium to manufacture nuclear fuel that can power commercial nuclear plants. The 34 tons of plutonium could power a nuclear plant for more than 34 years.

Members of the NRC toured radiological facilities at Los Alamos' Plutonium Facility, including ROBOCAL—the robotic calorimetry and gamma isotopic system. ROBOCAL is part of the Advanced Recovery and Integrated Extraction System (ARIES) and includes a gantry robot, a temporary holding and retrieval device, assay instrumentation, and computers.



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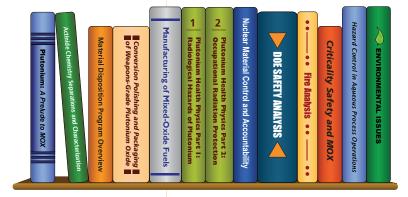
In the MOX cycle, the plutonium from a pit is removed and converted into an oxide powder, which is mixed with depleted uranium oxide powder and pressed under high temperatures into fuel pellets—a process called sintering. The pellets are sealed inside corrosion-resistant zirconium-alloy fuel rods, which are then bundled into fuel assemblies containing up to 300 rods each and shipped to commercial power plants.

Since the early 1990s, Los Alamos and Sandia National Laboratories, along with Savannah River, have been working on various aspects of making MOX fuels a reality in the United States. It's a technical challenge to produce the plutonium oxide to meet the required specifications for MOX fuels for commercial power reactors. Los Alamos produced the plutonium-oxide powder needed for MOX lead test assemblies as a first step in the U.S. program. In a joint venture, France then manufactured the MOX test assemblies at a plant in Marcoule in 2005 for use in commercial power a reactor in South Carolina. That reactor has been test-burning the fuel since the summer of 2005, and the fuel has performed as anticipated.

Still, the United States and Russia are years away from actually burning the 34 tons of plutonium in commercial reactors. After the test is finished, the United States will need to build its own MOX fuel fabrication facility and a facility to take the weapons-grade plutonium and convert it into a powder to produce fuel pellets. Those facilities are slated for the Savannah River Site in South Carolina.

Thirteen topics

The seminar for the NRL regulators covers a spectrum of topics, ranging from MOX production and operations to hazard control and environmental issues. About 70 people, primarily from the NRC, have attended multiple-day seminars at Los Alamos, the latest of which was held in August 2006. Several hundred people from various government agencies have attended half-day seminars at Savannah River. The Los Alamos seminar includes a tour of radio-



logical facilities and operations at the TA-55 Plutonium Facility (PF-4), which is home to the Laboratory's MOX operations. A brief overview of each section of the seminar is given below.

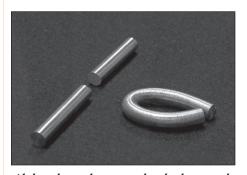


Staudhammer led off the seminar with an overview of the metallurgical properties of plutonium, the problems associated with its conversion to MOX fuel, the physical properties needed in the production of MOX fuel, and the quality-control parameters in the context of production and uniformity of product required for MOX fuels.

Plutonium metal presents a challenge to researchers because of its many unusual properties. Its six allotropic phases undergo large changes in volume during transformations from its stable room-temperature alpha phase to its liquid phase. The alpha, beta, and gamma phases exhibit large coefficients of thermal expansion, while the delta phase exhibits a negative coefficient of thermal expansion. Plutonium has a low melting point, its volume decreases upon melting, and it has poor oxidation and corrosion resistance. Plutonium also has the largest viscosity of any pure liquid metal and has an exceptionally wide range of liquid stability, from 640 to 3235 degrees Celsius.

The isotopic composition of weapons-grade plutonium is different from reactor-grade plutonium. Weapons-grade contains more plutonium-239, making it more fissionable, while reactor-grade plutonium contains more plutonium-240 and -241, making it more radioactive. Delta-phase plutonium is desirable for use in weapons because it is malleable, but it isn't stable at room temperature unless it is alloyed with another element, such as gallium.

However, excessive amounts of gallium can cause problems in a reactor fuel. At high concentrations, gallium can affect the sintering process when the mixed oxide is turned into a pellet. The temperatures required for sintering turn the gallium into a gas, which deposits back on the furnace walls as a metal. In addition, while gallium does not interfere with the fission process, it does chemically react over time with zirconium, the metal used in the fuel rods. The process within a reactor environment is still not completely understood.



Alpha-phase plutonium has high strength and no ductility; delta-phase plutonium is malleable but unstable at room temperature unless it has been alloyed with another element. The bar of unalloyed alpha plutonium snapped in half when it was bent, while the gallium-alloyed delta-phase sample bent easily. The photo was taken in the 1980s at TA-55.





British Nuclear Fuel Ltd.

A pressurized water reactor fuel assembly (top) and a storage facility for spent nuclear fuel.

Much of the solution chemical behavior of plutonium depends on the nature of its oxidation state. The metal ion in each of those states can form a variety of molecular complexes, each with a characteristic solubility, spectroscopic properties, and chemical reactivity. Chemists have mastered the ability to manipulate these oxidation states to afford efficient chemical separations of uranium and plutonium from all the other elements in spent nuclear fuel and recombine them for MOX fuel fabrication.

Actinide Chemistry Separations and Characterization

David L. Clark and Gordon Jarvinen, director and associate director of the Seaborg Institute, briefed the class on the chemical uniqueness of the actinides, their fission process, and their neutron-absorption ability to generate higher actinides. They also discussed processes used to recover plutonium and uranium, as well as general methods to prepare MOX fuel.

The discovery of the fission process during World War II resulted in an explosive expansion of the synthesis of new actinide elements and knowledge of actinide chemistry. One of the challenges in separations science is the efficient partitioning of the components of spent nuclear fuel. Fission products are dispersed atomically in the fuel matrix and include about 30 elements from all groups of the periodic table. The high radioactivity of the fission products requires the use of heavily shielded and remotely operated equipment to perform the separation operations.

One widely used separations process is known as PUREX (Plutonium—Uranium Reduction Extraction), which was developed in the late 1940s and early 1950s based on exploratory work performed during the Manhattan Project. PUREX is a liquid-liquid extraction process that extracts uranium and plutonium from the minor actinides and the fission products. It has been used to process thousands of tons of irradiated nuclear fuel to recover uranium and plutonium; in civilian energy programs the plutonium is often recycled to MOX fuel.

Clark and Jarvinen also discussed the Global Nuclear Energy Partnership (GNEP) out of the DOE Office of Nuclear Energy, which is evaluating alternatives to the once-through fuel cycle. GNEP is intended to expand domestic use of nuclear power, demonstrate more proliferation-resistant recycling, minimize nuclear waste, develop advanced burner reactors, establish reliable fuel services, demonstrate small-scale reactors, and develop enhanced nuclear safeguards. The DOE plan calls for three new facilities: a nuclear fuel recycling center to separate the spent fuel and produce fuel for an advanced fast reactor, an advanced fast reactor to burn the actinide elements and produce electricity, and an advanced fuel cycle research and development center to develop and demonstrate the new technology.



Material Disposition Program Overview

Steven McKee of the Nuclear Nonproliferation Program presented two topics. The first was a discussion of the U.S. surplus plutonium program to reduce the global nuclear danger by safely dispositioning tons of plutonium removed from dismantled nuclear weapons. He highlighted the methodology and programmatic requirements, including aspects relevant to Russia.

In 1998, the United States and Russia entered into a Scientific and Technical Cooperation Agreement that provides a mechanism for experts from the two countries to work together on plutonium disposition technologies. In September 2000, the two countries signed a Plutonium Management and Disposition Agreement (PMDA) that commits each to dispose of 34 metric tons of surplus weapons-grade plutonium. And in July 2006, the DOE and the Federal Atomic Energy, Russian Federation (Rosatom), reaffirmed their commitment to implementing the 2000 agreement.

But progress has been slowed for the past several years because there are risks involved and there was no liability agreement to protect either country in case something unanticipated went wrong, such as difficulties with using the weapons-grade plutonium in MOX in currently deployed commercial reactors. The United States and Russia cleared a major diplomatic hurdle in September 2006 that gets both sides closer to dealing with surplus weapons-grade plutonium through use in MOX fuel when the two countries agreed on liability protection for the United States so that it can help Russia with its part of the equation.

Conversion Polishing and Packaging
of Weapons-Grade Plutonium Oxide

Pit disassembly and conversion are the heart of the disposition program. McKee's second topic was an overview of a major part of the program: the Advanced Recovery and Integrated Extraction System (ARIES) in PF-4. ARIES is used to remove gallium and other impurities from the weapons-grade plutonium in a process called polishing—or final purification. The ARIES approach prepares weapons-grade plutonium for long-term storage and disposition in a form that can be quantifiably verified by nondestructive assay by the International Atomic Energy Agency.

The ARIES technology has been shown to reduce worker exposure and minimize waste. It has been demonstrated in operations that take place in six gloveboxes and one non-glovebox area in PF-4. With ARIES, the plutonium from a pit can be converted to an oxide form, the required form for MOX fuel, by heating it in a direct metal oxidation (DMO) furnace. Plutonium can also be removed using the hydride/dehyrdride process, which forms plutonium hydride powder, which is then thermally treated to form a plutonium metal puck. The pucks also can be converted to the oxide form by processing them through the DMO furnace.



A view inside a glovebox port of a plutonium-packaging robot on the Advanced Recovery and Integrated Extraction System (ARIES) line.

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After the plutonium has been converted, it is packaged to meet DOE long-term storage criteria in a series of three nested containers. Before the first sealed container is removed from the glovebox line, it is electrolytically cleaned to remove any contamination. The final can assembly is then robotically assayed to confirm the quantity of plutonium in each package—a measurement that is critical for nuclear security and safeguards.



MOX pellet manufacturing involves standard ceramic processing steps of milling, blending, pressing, sintering, and grinding, but in an extremely sensitive environment. The stringent production requirements and objectives to produce certifiable MOX fuels were discussed by Ken Chidester, retired from Los Alamos and now with Nuclear Fuels Technology Associates, LLC.

The MOX development program has three objectives: to improve fuel lifetime, to reduce personnel exposure during the fabrication process, and to improve fuel quality. MOX fuel must meet stringent federal nuclear quality assurance standards, which require rigorous sampling to guarantee a consistent high quality in the fuel pellets. Specifically, the final product must be a homogeneous blend of uranium and plutonium oxides with plutonium-oxide particles smaller than 10 microns and at concentrations of no more than 30 percent. There must be a batch-to-batch and pellet-to-pellet uniformity with even distribution throughout the rods, assemblies, and core because uneven distribution can result in hot and cold spots.

Manufacturing objectives also include controlling the plutonium throughout the process through in-line monitoring and inspection and using an internal recycling process to minimize waste. The final product must be purified to minimize neutron parasite absorbers and to minimize elements that could chemically interact with the cladding around the fuel pellets. Quality assurance and control play important parts in the process. Inspectors must keep a data record for each process step and produce a complete record of sample history as well as a final data package for each fuel batch and fuel-rod group.



Steve Costigan of the Radiation Protection Division presented two portions of the seminar on plutonium health physics. The first part covered U.S. and international regulations on radiation protection as well as dose variables, dose assessment, and risks from exposure, primarily for plutonium.

Four basic types of ionizing radiation are of concern in a radiological work environment: alpha, beta, gamma, and neutron. Alpha particles have a very low penetrating power and are not considered an external radiation hazard but can, if ingested, deposit large amounts of radiation in a small area of body tissue.



U.S. Department Of Energy/Science Photo Library

Gloved hands hold life-sized models of nuclear fuel pellets with their injection rod. These use fuel containing fissile uranium and plutonium surrounded by a core of nonfissile uranium-238. Photographed in 1979 at the Fast Flux Test Facility at the Hanford Site in Washington State.

Beta particles have low penetrating power and can be potentially hazardous to skin and eyes but cannot penetrate to internal organs or bone marrow. This type of external exposure is called "shallow dose." Beta particles, like alpha particles, can be hazardous when taken into the body.

Gamma rays and neutron particles have very high penetrating power and are external "whole body" hazards. The whole body—the area that contains the most radiation-sensitive organs—is the area from the top of the head to just below the elbow and just below the knee. Dose limits are lower for the "whole body" than for skin, extremities, and other organs. Gamma, or photon, radiation can be in the form of low-energy X-rays emitted from plutonium and americium; neutron radiation results from spontaneous fissioning and alpha particle collisions with certain light nuclei such as fluorine, oxygen, and beryllium.

Plutonium can deliver a significant internal dose in a single intake, but externally it is comparable to other low-level, chronic exposures of more-common chemicals. External and internal dose variables for plutonium include the age of the material, the isotopic composition process, and its chemical and physical forms. Internal dose variables also include the route of entry; human variables such as age, genetic predisposition, chronic factors (smoking, diet, previous chemical exposure, etc.); and treatment options for removing the radionuclide from the body

The organs that are affected and the severity of the radiation dose depend on two things: how the plutonium enters the body and how much was taken in. Inhaled plutonium is generally more hazardous than ingested plutonium because it is more readily absorbed into the bloodstream via the lungs than via the gastrointestinal tract.

Plutonium Health Physics Part 2:
Occupational Radiation Protection

Costigan continued his discussion of plutonium health physics with a look at the DOE regulatory drivers for radiological control and plutonium good practices and the As Low As Reasonably Achievable (ALARA) program. A MOX fuel facility will have to adhere to numerous DOE standards for occupational radiation protection as well as facility-, group-, and process-specific operation requirements.

A radiation protection program has many components, including but not limited to emergency response, dose standards, personnel dosimetry, training and qualification, access control, contamination control, radioactive material control, and reporting. Emergency response priorities must address personnel safety (spill, contamination, and material intake), environmental concerns (material release and waste generation), and facility concerns (programmatic

n, onium Paper Wood Lead Water Skin Plastic Steel Thick Concrete gen, Clothing Aluminum Concrete

Four basic types of radiation are of concern in a radiological environment.
Alpha particles can be stopped by something as thin as a piece of paper, while neutron particles can penetrate through lead and steel, but can be stopped (or reflected) by water and thick concrete.

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At Los Alamos, some 100 men and women work with plutonium routinely in the only remaining plutonium processing facility in the United States. From the moment they enter the facility, they follow rigid safety precautions and are protected by specially designed clothes, gloves, and goggles. Because all of the actinide elements are radioactive, special containment and shielded facilities are mandatory for safe handling of these substances. Workers use gloveboxes, like those shown above, for their research with actinides, handling even minute quantities with amazing dexterity.

impacts). Emergency response steps include evacuation, mitigation, notification, investigation, and recovery.

The ALARA program is a regulatory requirement for all radiation safety programs and is an approach used to minimize radiation doses and releases of radioactive materials. ALARA is based on the assumption that all radiation exposure carries risk and that the assumed risk of exposure to ionizing radiation must be balanced against the benefit of performing the work.

Plutonium presents unique technological challenges because of its high radiotoxicity, low concern thresholds (release limits), high mobility, the physics of alpha detection (its geometry and sensitivity), and interfering issues such as radon and gamma and neutron emitters.

To address these challenges, plutonium operations must make efficient use of engineered controls for containment (e.g., gloveboxes), confinement (e.g., hoods and open-front boxes), ventilation, filtration, and limiting exposure (e.g., shielding, remote handling, and access control); contamination control such as alarm systems and workplace monitoring; administrative controls such as procedures, training, and personal protective equipment; and instrumentation for air, contamination, and dose-rate monitoring.

Nuclear Material Control and Accountability

Dennis Brandt of the Program Management Division briefed the class on material control and accountability (MC&A) requirements governing the labeling, material movement, and control and application of tamper-indicating devices, as well as assay methods and analysis techniques. While Los Alamos was historically a research facility rather than a production-oriented facility, many of the same control and accountability requirements would be in place at a MOX fuel fabrication facility.

The integrated MC&A program at Los Alamos uses several methods to track nuclear material, including a batch-accounting system rather than campaign accounting, which results in a more timely and localized detection of losses; direct operator input to the accounting system; and reliance on process accountability flow diagrams. Other methods include continuous review and oversight by accounting staff, daily material balances, bimonthly physical inventories, and a shutdown, cleanout inventory once a year.

After the metal or oxide has been converted and packaged on ARIES, a series of nondestructive assay instruments confirms the quantity of plutonium in the package. Similar techniques will be used by international inspectors to confirm the contents without having to open the package. Nonassay methods in use at TA-55 include passive neutron coincidence counting, active neutron assay, segmented gamma scanning, calorimetry/gamma-ray spectroscopy, electronic balances, and gamma-based solution assay. Destructive analysis techniques such as mass spectroscopy are also used on elemental plutonium and uranium and trace elements.



Patrick McClure of the Safety Basis Division discussed how to conduct a process hazard analysis for a mixed-oxide lead test assembly (LTA) as well as how to identify potential hazards and the protective features that can be used to minimize hazards.

The proposed process for fabricating MOX LTA pellets is a series of four steps. The first involves the feed preparation where the plutonium oxide is produced in the ARIES line and then purified, or polished, through dissolution and ion exchange. The second step is the blending process in which 5-kilogram batches of master blend (30 percent plutonium oxide) are combined to form a 15-kilogram master blend batch, which is then blended with uranium oxide to product a production batch. Pressing the pellets comes next. The master

Material

blend is loaded using an auger and a vibrating feed trough into several die cavities per press cycle and the green pellets are dropped into a sintering boat. In the final step the pellets are sintered in an argon/hydrogen atmosphere, ground to proper dimensions, and transferred in trays for rod loading.

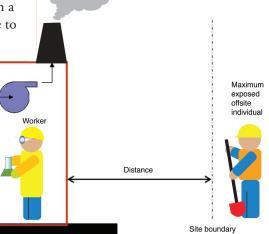
Potential hazards in the process steps have been identified. They range from a drop scenario such as a blend jar dropping to the glovebox floor, a

criticality event, a steam explosion, a furnace overheating, loss of inert atmosphere, a fire or seismic event, or a power failure. Protection features in place in PF-4 to mitigate potential hazards include the use of gloveboxes, the structure of the blender, the design of the ventilation systems, the use of HEPA filters and continuous air monitors, and the setting of criticality limits.



McClure presented a second section of the seminar on the history of fires in DOE nonreactor nuclear facilities, fire analysis, and modeling of such events to mitigate future scenarios. He also discussed PF-4 safety systems, structures, and components as well as technical safety requirements for future operations.

Two Rocky Flats fires were specifically discussed. The first occurred in 1957 in Building 71, a "fireproof" building with low combustible loading. The fire was caused by the spontaneous combustion inside a glovebox line of plutonium skulls (impure plutonium in a metal casting residue). The fire burned the Plexiglas glovebox windows and a series of combustible filters, resulting in

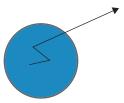


This illustration shows the safety controls in place in PF-4. Materials at risk are handled inside a glovebox, which is located inside a specially designed secure building. The building has special filter and ventilation systems and is set back a specified distance from the site boundary.

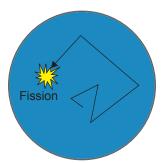
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Parameters that affect criticality safety:

Volume

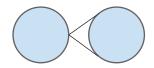


More likely to leak neutrons
Requires higher concentrations as
solutions or higher densities as
metals and oxides to reach critical.

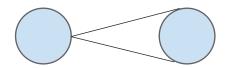


Less likely to leak neutrons
Requires lower concentrations or
densities to reach critical.

Interaction



More likely to interact



Less likely to interact

explosions from combustion gases. Firefighters first attempted to put out the fire with the conventional method: carbon dioxide extinguishers. It was feared that applying water would create a criticality event caused by the intense heat turning the water into steam, which would explode and send plutonium particles toward Denver. The carbon dioxide failed and firefighters had no choice but to use water. It worked, even though the fire burned for thirteen hours.

A second fire in 1969 in Buildings 776-777 started in a plutonium storage box. As in the 1957 fire, Plexiglas was ignited, as well as Benelex, a neutron-radiation shielding material made out of wood fiber and plastic. The fire spread throughout the glovebox ventilation line. And again, as in the 1957 fire, attempts were made to control the fire with carbon dioxide extinguishers, but firefighters soon switched to water. The fire was controlled within several hours, but the facility suffered extensive damage and contamination.

By analyzing the Rocky Flats fires as well as others, stringent regulations have been established for fire protection and fire suppression in nuclear and nonnuclear facilities. McClure noted the fire safety features in place at PF-4, including the exterior and interior walls built to withstand fires of several hours' duration, and a wet-pipe fire sprinkler system in the general areas and a dry-pipe system in the vault. Additional fire safety features include redundant fire water supply tanks and fire pumps, Halon systems, a fire-alarm system that includes heat detectors in gloveboxes and that activates the closure of fire doors, and low-combustible loading limits for gloveboxes and laboratories.

Criticality Safety and MOX

Shean Monahan of the Safety Basis Division discussed the fundamentals of criticality safety, acceptable safety margins and standards, and accident case studies as applicable to MOX production.

The critical mass of a fissile material is the amount needed to sustain a nuclear chain reaction. If the amount of fissile material is too small, some neutrons generated by fission can escape and the chain reaction doesn't become self-sustaining. Adding a reflector around the fissile material increases the efficiency of the reactions and thus decreases the amount of material needed to achieve criticality.

For example, the critical mass of uranium-235 metal is 50 kilograms (kg) bare, but less than 25 kg reflected. The critical mass of weapons-grade (95 percent plutonium-239, 5 percent plutonium-240) alpha-plutonium metal is 11 kg bare, 5.8 kg reflected; delta-plutonium metal is 15.5 kg bare, 8 kg reflected. The same holds true for fissile materials in solution. The critical mass of an idealized plutonium-water solution is 1000 grams (g) bare, 500 g reflected; a uranium-water solution is 1600 g bare, 800 g reflected. The plutonium and uranium in these cases is 100 percent pure plutonium-239 or uranium-235 and the reflector in both solutions is "thick" water—meaning a few inches of water surrounding the solution volume.

A variety of interacting parameters affect criticality safety, including mass, volume, shape and geometry, moderation, density, reflection, concentration,

poisons, enrichment, and interaction. Ensuring criticality safety involves all of these parameters at the same time, which can be extremely complicated.

Criticality safety is of great importance in the MOX process. Mixed-oxide fuels are a combination of uranium and plutonium oxides that have similar characteristics (neutronics, burnup, and heat transfer) to low-enriched uranium (LEU) in nuclear power reactors.

From a criticality safety perspective, the same considerations are in place for MOX processing as for LEU processing and fuel fabrication. Each of the parameters important for criticality safety need to be considered for the various operations that will be used to generate MOX fuel assemblies, including feed processing and fuel fabrication.

Criticality safety procedures and methodologies are detailed in a standard developed by the American Nuclear Society: ANSI/ANS.8.12-1998. The standard "provides guidance for the prevention of criticality accidents in the handling, storing, processing, and transporting of plutonium-uranium fuel mixtures outside reactors and is applicable to all operations involving mixtures of plutonium and natural uranium." The NRC must also conduct a detailed nuclear criticality safety analyses for specific operations.

Hazard Control in Aqueous Process Operations

Kent Abney of the Actinide Process Chemistry Group briefed the class on the rigorous standards that are required for facility design, hazards analysis and control, work implementation and authorization, and product specification. Fissile and chemical material processing facilities have both general industrial as well as unique hazards. Abney identified—in order of decreasing risk or likelihood of consequence—the hazards associated with handling fissile materials at PF-4: criticality, chemical, contamination, radiation exposure, and industrial hygiene.

The general approach to controlling hazards has five parts. The first two include defining the work and analyzing the hazards (including handling fissile material, chemicals, high temperatures or pressures). The third is implementing appropriate hazard controls (eliminate the hazard, substitute a lower-risk hazard, use engineered barriers like gloveboxes and ventilation, put in place administrative controls like training and integrated work documents, and use appropriate personal protection equipment). The final two are authorizing and performing the work; and analyzing and getting feedback from lessons learned.

He described several observations from analyzing criticality accidents in process facilities. Twenty-one of the 22 worldwide accidents from 1953 to 1999 occurred with fissile material in solutions or slurries. No accident occurred with fissile material during storage or transport, resulted in significant radiation exposures beyond the facility boundary, or resulted in radiation doses exceeding occupational limits.

Analysis of past criticality accidents has resulted in important lessons learned, among them that unfavorable geometry vessels should be avoided in high-concentration operations, the process should be well understood so that

Shape/geometry



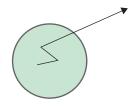
More likely to leak
Pencil tanks
(≤ 6" diameter)/slab tanks
and annular tanks/thin shells.



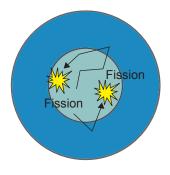


Less likely to leak
Solid spheres and cans with
height/diameter ≈ 1.0.

Reflection



Critical, bare sphere for uranium (93% enrichment) metal is about 50 kilograms.



Critical, reflected sphere for uranium (93% enrichment) metal is less than 25 kilograms.

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abnormal conditions can be identified, and criticality control and fissile material accountability should be integrated.



Harvey Decker of the Environmental Protection Division wrapped up the seminar with a discussion of the areas that must be addressed for a MOX production facility to succeed from an environmental compliance standpoint: environmental protection, waste management, certification and compliance programs, and quality assurance. These issues, if ignored, could affect the health and safety of the worker and the environment, delay work, affect regulatory compliance, and result in substantial liability costs.

Environmental protection reviews are generally performed following specific regulatory requirements such as the Clean Air Act for airborne emissions and the Resource Conservation and Recovery Act (RCRA) for dealing with wastes. The National Environmental Policy Act requires federal or federally funded facilities to evaluate all potential environmental impacts on water, air, soil, endangered species, and cultural and historic landmarks during the planning stages of a project.

An air-quality review must consider generators and other combustion equipment, exhaust systems, activities involving radionuclides and beryllium, solvent-cleaning operations, storage tanks, and open burning and excavation. A water-quality review will look at generation and discharge of waste- and potable water, wastewater treatment and collection, septic and oil-storage tanks, and storm-water runoff.

Waste management is an essential element to any production facility, whether the waste is radioactive, hazardous, mixed, sanitary, industrial liquid, salvage, or recycled. A waste-management program will be concerned with the characteristics of the waste—its type and level of radioactivity, hazardous constituents as determined by RCRA, and safe handling issues. The program will also look at regulatory compliance requirements, including federal, state, and local; disposal paths for waste by-products; waste treatment, minimization, and pollution prevention; process and facility designs; packaging and transporting for disposal; and potential waste recycling or reclamation methods.

Regulatory-compliance issues are concerned with waste minimization, generation, characterization, segregation, storage, treatment, packaging and transportation, and disposal. Certification, compliance, and quality-assurance programs are governed by DOE Orders and any federal, state, or local operating permits a facility may be required to have.

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The challenge within the Seaborg Institute has always been to respond to the Laboratory's eminent role in supporting the Untied States' needs to further the understanding and safe deployment of our nuclear potential The MOX seminar encompassed all of these aspects in a unique way.

The design of a MOX facility must address a myriad of environmental compliance issues. The bottom line is to protect the health and safety of all creatures great and small.



AL GHIORSO RECALLS A MOMENT IN TIME



The following article is based on a talk given by Al Ghiorso of UC-Berkeley at the National Atomic Museum's "Einstein Group" in Albuquerque in March 2006. While the immediate connection to Los Alamos was minimal, Ghiorso's talk recounts a bit of largely undocumented history that led to Los Alamos' work in developing the hydrogen bomb. ARQ thanks Al Ghiorso, Tom Keenan, Robert Penneman, and George Cowan for contributing to this article.

FOUR PROPHETIC DISINTEGRATIONS PER MINUTE LED TO THE DEVELOPMENT OF THE HYDROGEN BOMB

At one time or another, most of us have heard the "tick-tick-tick" of a radiation counter. We might have heard the sound at work, or, if we didn't normally deal with radioactive materials, we might have heard the sound in a science-fiction movie: "Gee willikers, listen to that counter . . . there must be an *atomic bomb* hidden in the building!"

Usually, the audio "tick-tick" reflects several hundred or possibly several thousand "events" per minute. A single tick sound comes from the spontaneous disintegration of a radioactive nucleus; numbers of nuclei can easily be in the tens or hundreds of millions, leading to many audible disintegrations per given unit of time (usually one minute).

However, there was one instance in our recent history when a very, very few number of ticks had extraordinary consequences. We're not talking about thousands or even hundreds of disintegrations; we're talking about approximately four disintegrations per minute—or averaging one every 15 seconds.

To set the stage, we have to go back to the late 1940s when only the United States held the techniques and knowledge to manufacture a nuclear weapon. Or so we thought.

In the late summer of 1949, air-borne radioactivity was picked up on filters aboard U.S. Air Force B-29 reconnaissance aircraft while flying routine missions near the Siberian borders. These planes were on patrol to look for evidence of Soviet bomb tests. This radioactivity indicated that some sort of nuclear event had taken place.

The filter samples were brought back to the United States and analyzed in California. Some fission products were detected, including barium-140, molybdenum-99, cerium-144, lanthanum-140, and yttrium-91. All of these had modest half-lives (i.e., days to months). Their distribution was the first hint that the Russian "event" was probably a weapon.

In a bomb or an instantaneous explosion, the fission products are generated in a fixed, known distribution. In a reactor, the fission process continues over an extended period of time. Short-lived fission products decay and long-lived products accumulate and predominate. The observed distributions were

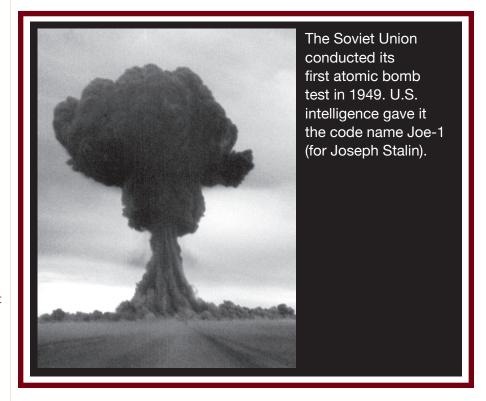


Michael Crescenti National Atomic Museum

Dr. Albert Ghiorso received the National Award of Nuclear Science at last year's Ninth Annual Einstein Society Gala at the National Atomic Museum in Albuquerque. The award is presented to a person who has had an impact on nuclear issues. It celebrates the wide scope of achievement and commitment to furthering scientific endeavor made by individuals in areas of military leadership, medical technology, public policy and government, energy sciences, education, and space exploration.

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told that some members of the Truman administration were sure that what had been detected was more likely a runaway nuclear reactor accident than an atomic bomb explosion. This was because the CIA was adamant in claiming that it was impossible for Soviet scientists to have constructed a bomb as quickly they had. That is why, when President Truman announced that there had been a "nuclear explosion" in the Soviet Union, he carefully did not say that it was a bomb. On the other hand, the chemists who analyzed the fission-product debris were certain from their decay measurements that an atomic bomb had to be its origin. Confirming evidence was needed. Was it a bomb or not?



consistent with instantaneous formation at a particular moment in time, which corresponded to the presumed time of the test.

The radioactive decay showed that they were fission products that had been freshly produced and thus could not have arisen from a nuclear reactor accident. These determinations made the debris interesting, but more material was needed to increase the power of the analyses by examining its plutonium fraction. Fortunately, the Naval Research Laboratory (NRL) had collected a much larger sample of the cloud as it passed over Washington, D.C., dropping rainwater into giant collectors on the roof of the NRL building.

The rainwater was chemically concentrated to produce a significant heavy-element fraction and sent to Los Alamos via special armed courier, where it was found to contain three alpha counts per minute of plutonium. This in itself did not prove that the explosion was an atomic bomb because a uranium reactor would also contain plutonium even if it had been operating for only a short length of time. The important definitive measurement needed was the isotopic composition of the plutonium. In particular, how much plutonium-238 was present?

At the time, experts could not imagine that the Soviets had the scientific know-how to make a workable bomb in spite of the information spies had transmitted during and just after World War II. Even Gen. Leslie Groves, who headed the Manhattan District Project Y at Los Alamos, estimated that it would be at least the mid-1950s before the Soviets could even assemble a prototype.



No one believed that the Soviets had detonated a deliverable weapon; some people felt that a reactor might have exploded or gone supercritical as did the Chernobyl reactor many years later. Indeed, the Soviets somewhat encouraged this conclusion in that their sketchy news releases mentioned a reactor "accident." President Truman, at the suggestion of his political advisors, only announced that there had been a "nuclear explosion" in the Soviet Union; the word "bomb" was deliberately not used.

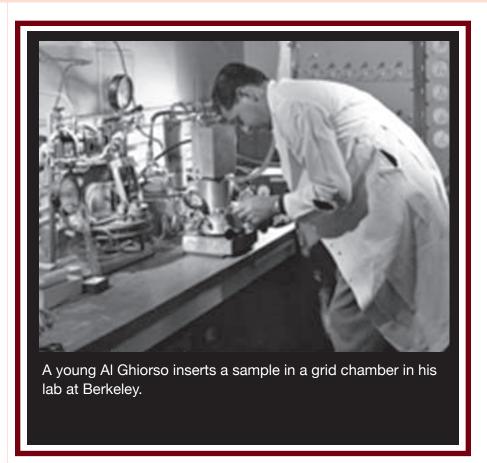
In the late 1940s, the only AEC laboratories with experience in plutonium chemistry were Los Alamos; Berkeley, California; and Argonne (near Chicago). Livermore did not exist then.

One of the best ways to find out what kind of plutonium was in those samples was to analyze the energy of the emitted alpha particles. When plutonium disintegrates, or decays, it emits an alpha particle or a helium nucleus of a specific energy. Ordinarily, these are fairly easy to detect, and even in the 1940s there were instruments available to measure the energies of the emitted alpha particles. However, those instruments were most often used when thousands of disintegrations were taking place (remember the "tick-tick-tick"), which was certainly not the case with the sample that emitted only about four disintegrations per minute.

At Los Alamos, a special radiochemistry group under the leadership of Rod Spence had been formed specifically to study and evaluate the emissions from AG: (The first evidence that an atomic bomb had been detonated by the Soviet Union came from aircraft that had picked up explosion debris from the airborne cloud while flying routine missions near the borders of Siberia. These planes had been patrolling since spring 1949 and were operated by AFOAT-1, a newly formed Air Force group that was tasked specifically to look for evidence of Soviet bomb tests. How does one tell the difference between an atomic-bomb explosion and a runaway nuclear reactor explosion? The definitive way is to look for plutonium-238. Physicists knew that plutonium-238 would be made by fast neutrons in a nuclear explosion by the n, 2n reaction, whereas in reactor-produced plutonium, fast neutrons are slowed down by the moderator and the relative number of plutonium-238 atoms that would be produced relative to plutonium-239 would be fewer by about a factor of a hundred. However, since the half-life (about 87 years) of plutonium-238 is about 300 times shorter than that of plutonium-239, it should be readily detectable by measuring its alpha activity even if the amount made is very small. The problem was that at that time there was no instrument available that could make this measurement. Or was there?

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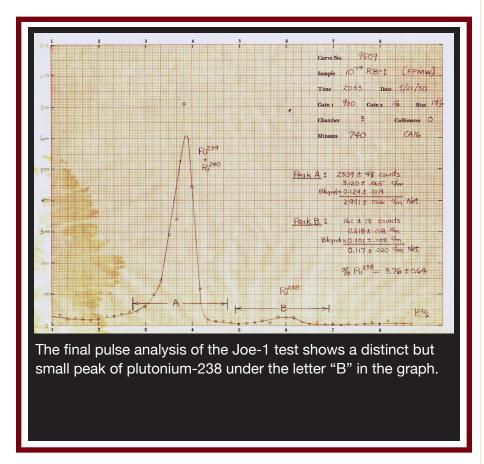
AG: CAbout six months after the Joe-1 incident, the discovery of elements 97 and 98, berkelium and californium, in Berkeley was published by the Seaborg group, of which I was a member. This work called attention to the fact that we had developed an enormous sensitivity for analyzing small amounts of alpha activity. These new elements had been made by bombarding microgram quantities of americium and curium by beams of helium ions to make small amounts of alpha activity coming from elements 97 and 98 in different bombardments. My role in the team had been, and would continue to be, to devise and use suitable instrumentation that would be needed for the work with the heaviest elements. I had found from the literature that what was available was meager and inadequate, so I had to invent and develop new methods. After a period of years these techniques had been honed to fit the goals set by the search for heavier elements, and thus the program came to depend completely on our 48-channel pulse-height analyzer.



atomic bomb tests. Someone knew (or had heard) that Los Alamos could perform alpha energy analyses. However, the one alpha energy analyzer on the Hill belonged to Bob Penneman's group. Spence and Penneman quickly realized that this analyzer would not be able to do the delicate and sensitive job that was dictated by the small sample size, but Pence knew that Al Ghiorso at Berkeley had a much more sophisticated device, which was better suited to the problem at hand.

Ghiorso was part of Glenn Seaborg's Berkeley team that was busy "discovering" new elements; by 1949, they had isolated elements through Atomic Number 98 (californium). Part of their success was because of the successful electronic analyses of alpha energies at extremely low disintegration rates (as in disintegrations per minute, not seconds) in devices designed and built by Ghiorso. He was an electrical engineer and not a chemist, but his advances in delicate electronics made him a key member of Seaborg's group. There were no solid-state circuitry or transistors in 1949, so Ghiorso's instruments employed state-of-the art vacuum tubes, resistors, and capacitors. Counters to record the data were mechanical.

The sample was sent from Los Alamos to Berkeley (again under armed escort), and Ghiorso went to work. A sample this small dictated that many



hours of "counting" would be necessary to obtain a statistically valid number to analyze. Ghiorso was able to show that the sample contained not only plutonium-239 but that a few percent of the activity was due to plutonium-238. This isotope comes from a reaction of plutonium-239 with energetic ("fast") neutrons. If plutonium-239 is hit by a fast neutron it momentarily becomes plutonium-240, but rapidly emits two neutrons to transform back to plutonium-238. In nuclear shorthand, this is written as $Pu^{239}(n,2n)Pu^{238}$.

Ghiorso knew that the plutonium-238 had to come from a weapon because plutonium bombs have fast neutrons associated with them; reactors do not. Thus, the sample represented a plutonium bomb event and not a reactor explosion. Ghiorso's data were buttressed by analogous measurements from the 1945 Trinity Test. The copy of Ghiorso's original trace clearly shows the distinct—but small—peak of plutonium-238 at the bottom of the graph under the letter "B" in the image shown above.

When these data were transmitted to Washington, they attracted immediate attention. The fact that the fission products with various half-lives had originated at the same moment (i.e., a bomb) rather than over some time (i.e., a reactor) provided corroboration of the plutonium-238 data. They were accepted

AG: **C** The Soviet A-bomb sample emitted less than 200 alpha counts per hour, so I had to run it many times to determine the plutonium-238 content with a reasonable statistical accuracy. Final results of the Joe-1 test measurements were 5.15 MeV plutonium-239, -240 and 2.959±0.022 net counts per minute; 5.50 MeV plutonium-238 and 0.149±0.007 net counts per minute; and plutonium-238 total activity was 4.8 percent. The ratio of plutonium-238 to -239 that I measured was roughly half that produced in the Trinity Test. In contrast, the Joe-2 test that I measured in October 1951 showed an efficiency that was comparable to or better than that of Trinity. I understand that the first (Joe-1) was a carbon copy of the U.S. device—its design obtained from spies—whereas the second Soviet test was an original Soviet design. My measurements showed that both devices used plutonium-239. 77

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of the Joe-1 incident, not only because of the intensity of the work associated with the discoveries of elements 97 and 98, but also because of the level of secrecy that was imposed around the possibility that the Russians might have exploded an atomic bomb. In May 1950 I was approached by Roderick Spence, the head of the Los Alamos team that was analyzing the Joe-1 activity. It never occurred to me to connect his request with the Soviet explosion as published in the news media. Spence merely asked if I could analyze a small sample (only 3 alpha counts per minute) saying that the measurement was important for national security. He said that it was related to a highly classified matter that could not be divulged to me since I did not have the necessary security clearance. I agreed to make the measurement, and the sample was sent to me by a special courier. I knew that it must be very special indeed if it had to be hand-carried from Los Alamos by a security officer. I inserted the small platinum plate into the grid chamber and started to collect data. After a few hours I knew where the sample had come from because it was very much like one that I had measured in 1945 when I was in Chicago.



An early badge photo of Roderick Spence, who was the leader of a special radiochemistry group at Los Alamos that had been formed to study and evaluate emissions from atomic bomb tests.

and President Truman, at the suggestion of his science—not political—advisors, soon directed that Los Alamos go on a mandatory six-day workweek with an all-out effort to develop a hydrogen bomb—or Super as it was then called. The Russian "event" that initiated these efforts was given the code name Joe-1 for Joseph Stalin. In just three years, the Los Alamos efforts culminated in the successful George and Mike tests to demonstrate nuclear fusion.

In notes to his talk, Ghiorso reminisced about being asked to come to Washington to report on what he had found out about Joe-1. "As far as I can figure our from Google at this late date, the committee that I reported to was probably the Committee on Atomic Energy of the Joint Research and Development Board of the Armed Services. Robert Oppenheimer was its chairman and he took a personal interest in developing techniques for detecting foreign atom bomb explosions.

"Unfortunately, I do not remember anything about the hearing itself," his notes continue. "I found it fascinating to play the role of detective to find out what had happened on the other side of the world. It may have been because of the very hush-hush nature of this work that I did not document what happened subsequently. I can't recall other names or details, unfortunately; I was only 35 and probably terrified!"

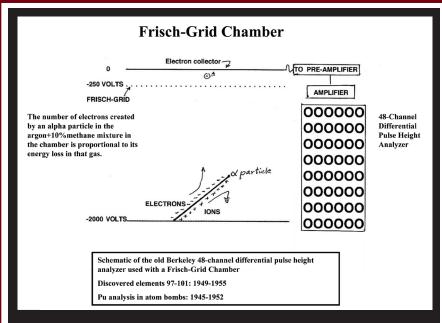


AG: The sample that I had measured in 1945 had come from the Trinity Test. I deduced that the new sample I was testing must have originated somewhere in the Soviet Union.

Flashback to the Metallurgical Lab in 1945: The Los Alamos bomb physicists wanted to determine the number of neutrons created in the Trinity explosion by measuring the amount of plutonium-238 that was produced by the n,2n reaction on plutonium-239. They came to Chicago to use the thermal column of the old CP-1 reactor that had been set up at a nearby site. Their plan was to use its thermal column to measure the number of slow-neutron-induced fissions of plutonium-239 in their explosion sample and compare it with that of the original plutonium. The difference in the fission/alpha ratios would be due to the alpha activity of plutonium-238, but because several uncertain corrections had to be made it was not a straightforward measurement.

When they heard about our newly completed 48-channel pulse height analyzer (in 1945 multichannel devices were not yet common) they contacted me to see if I could help them by measuring the plutonium-238 content directly with my alpha pulse analyzer. That sample contained hundreds of alpha counts per minute, so it was easy to make the measurement that they needed and I made the analysis quickly. That is how I came to know the typical signature of the alpha spectrum of plutonium after an atomic-bomb explosion. The amount of plutonium-238 formed is approximately proportional to the efficiency of the bomb, which means that the ratio of the alpha peak at 5.5 MeV (plutonium-238) to 5.16 MeV (plutonium-239 + -240) gives a good idea of the efficiency of the explosion. All of this took place some four years before the Joe-1 explosion. (Unfortunately, I could not find my original pulse analysis of the Trinity Test. I remember that the plutonium-238 intensity was close to 10 percent by activity of the total alphas.)

AG: (The device used for this research was a copy of the first instrument that we had constructed at the Metallurgical Laboratory at the University of Chicago. It used an array of 48 old-fashioned biased thyratron detectors arranged to measure the pulse height of each detected alpha particle. It had been vastly improved at Berkeley by the substitution of a gridded alpha particle ion chamber for the ordinary ion chamber that I had used at Chicago. With this system it was possible for us to measure accurately the energies of the alpha particles that were emitted from a thin sample. Its sensitivity was such that it could make accurate measurements that were able to differentiate among the various alpha-emitting isotopes in the heavy-element region. At that time I think that this facility was the only one in the world that was capable of analyzing very weak sources that might not emit more than an alpha count per hour, and it was the key tool in the discovery of elements 97 through 101. >>



A schematic of the Berkeley alpha grid chamber. A Frisch grid shields the collecting electrode from the moving positive ions. Even though only the electrons are collected, the movement of the + ions can be detected on the collecting electrode if the grid is not used. The grid prevents that and increases the resolution of the alpha particles. To make sure that all of the electrons are collected, the voltage on the grid is carefully adjusted to obtain the maximum pulse height.

$Pu^{239}+n \rightarrow Pu^{240}+n \rightarrow Pu^{241}$

Ghiorso's plutonium-241 idea.

AG: While pondering the problem, I thought of an additional measurement that I could make that would show how long the reactor had run to produce the plutonium in the bomb; why not measure the radioactivity from the plutonium-241 that also must be in the material? That isotope, with a half-life of 14 years, decays by emitting beta particles (electrons) with a very low energy. These electrons could not penetrate through the window of the usual Geiger counter, so I decided to try measuring the number of betas directly by using a windowless proportional counter, i.e., one where the sample is placed inside the counter. I decided that since the sample had been highly purified it was probably free from fission products. This seemed to work out very well; now I was able to measure how much plutonium-241 there was in the sample—and thus roughly an estimate of the amount of plutonium-240. The plutonium-241 isotope is made by successive neutron captures in the reactor ($Pu^{239}+n \rightarrow Pu^{240}+n \rightarrow Pu^{241}$). The amount of this isotope depends on how long the reactor had been operating before the uranium was removed from the pile to be processed. I roughly calibrated the counter's efficiency to count the beta particles from plutonium-241 by measuring the amount in samples of plutonium of various "ages" that had been recovered early on as the reactors came up to full power. When I made this comparison it showed that the amount of plutonium-240 was between 2.2 and 3.3 percent in the Joe-1 sample if it was produced in the same way as the American reactors. It showed that the reactor where it was made had operated for about a year.

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THE HUNT CONTINUES

Al Ghiorso's career as an element hunter dates back to the early 1940s when he and his colleagues discovered americium and curium (elements 95 and 96, respectively). His trophy wall now includes a dozen, including elements 97 through 106: berkelium, californium, einsteinium, fermium, mendelevium, nobelium, lawrencium, rutherfordium, dubnium, and seaborgium.

In the decades since Ghiorso discovered his first element, researchers have been hunting for more of the elusive prey. Last fall scientists from Lawrence Livermore National Laboratory and Russia's Joint Institute of Nuclear Research announced the creation of a new super-heavy element, number 118. The element was produced by bombarding a target of californium-249 with a beam of calcium-48 atoms. The new element has an extremely short lifetime of a few thousandths of a second and so far only three atoms of it have been created.

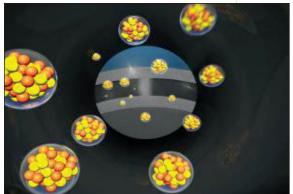
The element, currently known as ununoctium, has yet to be confirmed. "The process has to be done independently by another group, and the resources required are considerable," said Ken Moody, lear chemist at Livermore. "I would not be surprised if it took a few more

a nuclear chemist at Livermore. "I would not be surprised if it took a few more years before anybody tried."

The discovery of element 118 had been announced once before in 1999 by another research group, but because of accusations of falsified data, the claim was withdrawn.

Moody reports that one of his team's earlier results has recently been confirmed by Gesellschaft für Schwerionenforschung (GSI) in Germany. GSI reproduced Livermore's result for element 112 isotopes formed in the irradiation of uranium with calcium ions. In conjunction with recent radiochemical work performed by a Swiss group at the same laboratory in Russia where Livermore researchers perform their experiments, Moody and his colleagues hope that the eight-year-old result on element 114 will soon be accepted by the international community.

"We wrapped up the first phase of an experiment on element 120 in April," said Moody. "No candidate events have been found a this time, although we are still examining the data. We hope to resume next year because we can push the sensitivity of the experiment somewhat further."



Lawrence Livermore National Laboratory

An artist's conception of the experiment that produced element 118, in which a target of californium-249 is bombarded by a beam of calcium-48 atoms.

END NOTES

MIGRATION '07 CONFERENCE

The 11th international conference on the Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere (known as MIGRATION '07) will be held in Munich, Germany, August 26-31. David L. Clark, director of the Los Alamos branch of the Seaborg Institute, is a member of the international steering committee.

The MIGRATION conferences provide an international forum for the exchange on information on chemical processes controlling the migration behavior of actinides and fission products in natural systems. The information generated from the MIGRATION conferences is the basis for the fundamental understanding of the migration behavior of long-lived radionuclides, which is essential for the longer-term performance assessment of nuclear waste disposal.

Conference sessions will cover aquatic chemistry of actinides and fission products, migration behavior of radionuclides, and geochemical and transport modeling. A special session on organic matter modeling will be arranged with organizers of the 1st International Workshop on Organic Matter Modeling, held in Toulon, France, in 2005.

The conference is organized by Forschungszentrum Karlsruhe and the Institut Für Nukleare Entsorgung. Further information can be found on the conferences web site: http://www.fzk.de/migration2007.

ARQ WINS TWO AWARDS

Actinide Research Quarterly has won two awards in the 2006 Society for Technical Communications (STC) Southwest Regional Competition. ARQ won an Award of Excellence in technical publications and an Award of Merit in technical art. The awards were for three issues: "Postdoctoral research programs fuel the research engine," 2nd Quarter 2005; "Correlation effects in actinide systems," 3rd/4th Quarters 2005; and "Rocky Flats: From Weapons to Wildlife," 1st/2nd Quarters 2006.

The awards were presented to editor Meredith Coonley, illustrator/designer Kelly Parker, and scientific advisor David L. Clark, representing the Seaborg Institute. Coonley and Parker are with the Information Resource Management Division.

ARQ has won nine technical communication awards from STC over the past few years: six Awards of Excellence and three Awards of Merit.



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END NOTES

Lab photographer Dixon Wolf took this time-exposure night shot of a truck leaving the excavation dirt area across from the construction site for the Radiological Laboratory Utility Office Building, also known as the Rad Lab, at TA-55. The construction is Phase 1 of the Chemistry and Metallurgy Research Replacement (CMRR) Project, which will relocate mission-critical projects such as analytical chemistry, materials characterization, and actinide research from the decadesold CMR Building at TA-3.



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